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#### JP62278770

**Publication Title:** 

RECOVERING METHOD OF CO2 AND H2 FROM CONVERTER GAS

#### Abstract:

PURPOSE:To increase utilization by treating converter gas with a pressure swing adsorber (PSA), treating again with a PSA installed in series, and supplying the obtained H2 to a fuel cell to generate electric power and recovering exhaust heat as steam.

CONSTITUTION:LD converter gas is modified to H2, and treated with a pressure swing adsorber (PSA) 3 through a absorbin tower 1, and offgas RH2 which contains other gasses in addition to H2 is stored in a holder 4. The offgas RH2 is treated with a PSA 6 installed in series to obtain H2. The H2 obtained is supplied to a negative electrode 13 of a fuel cell 15 and reacted with oxygen in a positive electrode 14 to generate electric power. Exhaust gasses from the negative electrode 13 and the positive electrode 14 are supplied to a combustor 18 to completely burn and exhaust heat is recovered in a boiler 19. Therefore, electric power, steam, and fuel gas can be obtained from offgas from purification equipemnt of converter gas, and utilization is increased.

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母発明の名称

転炉ガスからのCO2, H2回収方法

②特 願 昭61-120218

**愛出 願 昭61(1986)5月27日** 

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#### **明 細 但**

1. 苑明の名称

転炉ガスからのCO1,H1回収方法

2. 特許商求の範囲

転炉ガスを変成し、次いで該変成ガス中の
COIを吸収塔にて吸収し、減吸収液よりCOIを
取得し、次いで、該吸収塔オフガスを昇圧し、
ア
正ガスをPSA(圧力式吸着装置)にて処理する
ことにより、COI及びHIを回収する方法にわい
て、該PSAオフガスを、更に直列に設けたPSA
Aにて処理した後に燃料電池に供給し、電力を取得し、該燃料電池のオフガスを完全燃焼せめ、
燃焼熱をスチームに変換利用することを特徴とする
転炉ガスからのCOI、HI回収方法。

3. 発明の詳細な説明

〔産業上の利用分野〕

本発明は、LD転炉等から製鋼時発生する転炉ガスからのCO。、H。を回収し、熱及び電気エネルギーを取得する方法に関するものである。

〔従来の技術〕

近時、製鋼用し口転炉等においては、便業時発生する、CO。CO。及びHiを含有する転炉供が スを積極的に個収し有効利用を図っている。

これら転炉ガス成分は、容別基準(以下同じ) にて、CO.10~15%, H.1%, CO65~75%, N.10%, O.1%である。

ての転炉ガスは、それ自体は燃料ガスとして使用しうるものであるが、更にこのガスを原料ガスとして、これに水蒸気を添加しシット反応を生せしめ、二酸化炭素及び水梁とし、付加価値を向上せしめる回収方法が一部に行われている。

第2回は、LD転炉ガスから、CO,及びH,を 回収するための従来法の工程圏である。

図において、前記組成のLD転炉ガスを原料として、該ガスを9~20㎏/clに昇圧し、これに水 蒸気を添加しシフト反応を起せしめる変成工程を 軽て、ガス成分を変成せしめる。(なお以上につ いては図示なし。)

次いで吸収塔1に変成ガスを導入し、吸収液と しての炭酸加里液 ( K . C O .) 中に、変成ガス中 のCO\*を吸収せしめ供配水素加里(KHCO\*) とし、これを水蒸気にてストリッピングし、吸収 被はK\*CO\*とし、一方ストリッピングされた CO\*ガスを製品とする。

COse 政出し、H s リッチの吸収塔1のオラガスは、PS A コンプレッサー2にて9~20kg/cd から9~25kg/cdに昇圧し、PS A (Pressure Sping Adsorption圧力式吸塑装置) 3にて処理する。PS Aにては、水素以外の気体を、圧力整を利用して吸激し、製品再純度水素(PHs) とオフガス(R Hs) とに分離し、オフガスはR Hs ホルグー4に貯留し、ブローワー5にて、他のプロセスの燃料用として送気している。

この場合のオフガスの成分例は、C O 12%, H 158%, C O 3.5%, N 126%, O 10.5%である。 [発明が解決しようとする問題点]

級上の如く、転炉ガスからCO\*, H \*を回収する従来の回収方法におけるオフガスは、H \*を約60%程度含有しているにも拘らず、単に燃料用としてのみ利用されているに過ぎない。

本発明の転炉ガスからのCOI, Hi回収方法においては、水素を約60%含有するPSAオフガスを更に直列に設けたPSAにて処理し、CO成分の少ない、濃度の高いHiを収率負く回収することができたので、燃料電池の燃料ガスとして供給を可能とし得て、

更に、燃料電池のオフガスを完全燃焼せしめその排料をスチームとして回収でき、電力、スチーム, 燃料ガスなどの多様エネルギーを一挙にかつコスト的に有利に回収することを可能とするものである。

製鉄所用辺の内院あるいは事務所等においては、スナーム、電力と共に始為、冷理問設備等が必要である。現状においては、前途の如く、転炉ガスからの値収COェ、Ha等の有力なエネルギー部があるに向らず、異電々力にて前途の様な福利、管理設備の需要をみたし、また給湯、冷観概には重油等を使用しているケースが多い。

本苑明は、転炉ガスからの C-O , . H , 製造設備の比較的カロリーが低い(500~1700KC \* ℓ / N ㎡)が、水素混度の高いオフガスを効率的に回収し、かつその保存するエネルギーを効率的に利用する方法を提供することを目的とするものである。

(問題点を解決するための手段)

本発明者等は、従来法における水素約60%を含有しているPSAオフガスに着目し、このオフガスの有効利用を研究し本発明に至ったものである。即ち本発明は、

転炉ガスを変成し、次いで、変成ガス中の CO。成分を吸収塔にてK。CO。液中に吸収し、 該吸収液より製品でO。を取得し、次いで該吸収

次に実施例について述べる。

#### (灾旋例)

第1回は、木発明の転炉ガスからのCOI, HI 回収方法の工程説明図である。

第1図において、1~4は前述の第2図と何ーなので説明を省略する。

6 は 直列 に 設け た P S A 、 7 は 真空 プローワー、 8 は ブローワー、 9 は バルブ、 1 0 は 熱交換器、 1 1 は 燃焼用 空気 ブローワー、 1 2 は 燃料電池、 1 3 は 水楽極室 (负極) . 1 4 は 酸素極室 (正極) 、 1 5 は電解液、 1 6 は ポンプ、 1 7 は予熱器、 1 8 は 燃烧器、 1 9 は ボィラー、 2 0 は 煙突 で ある。

第1 図に基づいて、本発明方法について述べる。 L D 転炉ガス (H :: 0.7%, C O: 69.8%, C O :: 14.7%, N :: 14.3%) 1.298 N m / H を 原料ガスとして、とのガスを 9 ~ 11kg / cd にコン プレッサーにて昇圧し、次いで、このガスを水蒸 気を用いシフト反応により水楽に変成し次いで吸 収場1にて、K : C O : 液によりガス中のC O: を 吸収し、吸収液を水燃気にてストリッピングし、 製品CO1を作る。次いで吸収塔1のオフガスを PSAコンプレッサー2にて16kg/㎡に昇圧しPSA3にて処理する。

PSA3にては、塔内の設理剤にH。以外のガスを吸着せしめ、製品PH。と、H。以外のガスをも含むオフガスRH。とに分解する。

オフガス·R H 』は一旦ホルダー4に貯留し、パルブ 9 を初如することにより、直宜プローワー8 (2000n a 水柱) にてオフガスを次のPSA6にて処理する。

PSA6からの処理水素ガス (553Nm²/H)は、PSA6にて処理したためにH。は89.5%, COは1.1%, CO。は0.4%, N。9%を含有し、 燃料電池に供給しうろものである。

次に燃料電池から電力及び蒸気を値収する実施 例について述べる。

上記PSA6からの処理水素ガスは真空ブローワー 7 にて吸引し、熱交換器10にてオフガス温度を、燃料電池12のオブガスの燃焼排熱を回収

似し水酸イオンと酸素になり、この酸素は再び電 毎反応に利用される。従って正極の反応は

12 0 1 + H 1 0 + 2 e - → 2 0 H -

この正極 1 4 で生成した水酸イオンは、電解板 1 5 中を移動して負極 1 3 に達し、全体の回路を 形成し電力 7 4 0 KWを取得した。

所かる燃料電池の負極13及び正極14からの 夫々190℃オフガスは燃焼器18に導入され、 完全燃焼し、430℃の燃焼排ガスは、ポイラー 19にてその廃熱を回収し6㎏/㎡の飽和蒸気 0.7T/Hを回収した。

なお負極13空内には統水の予熱器17を設け 予熱後ポイラー19に供給する。

ボイラー 1 9 のオフガスは熱交換器 1 0 にて前述の如く燃料電池への供給ガス及び空気を予熱後 歴突 2 0 より排煙する。

以上の結果、転炉ガスの評価が従来法に比して1.86倍の価値となった。

(発明の効果)

本発明の転炉ガスからのCO:, H,回収方法に

後の脱ガスと結交換し90℃に昇盛し、燃料電池 12の水条版(負極)13に導入する。

一方空気 2000 n m / H を ブローワー 1 1 に て 熱 交換 20 1 0 に 供給 し、 1 2 0 ℃ に 昇温 し 該空気 を 燃料 電池 1 2 の 粒素 揺 1 4 に 導入する。

燃料電池 1 2 は、リン酸水溶液 1 5 を電解液とし、これを挟んで、白金,ルチェウムからなる 2 つの電極から構成されている。

負極の水器電極 1 3 では、導入された水素ガスが、電極内の細孔を通って反応帯域近くに達し、電極に添加されている触媒に吸着され、活性な水素原子となる。この水素原子は次式のように、電解液中の水酸イオンと反応して水となり、その際 2 個の電子を電極に送る。

 $H_{3} + 2 O H^{-} \rightarrow 2 H_{3}O + 2 e^{-}$ 

正極の酸素値14では、触媒の存在のもとで、 電極から2個の電子をうけとり、外部から供給された空気中の酸素分子が、電解液からの水と反応 して次式のように、過酸化水素イオンと水酸イオンを生成し、過酸化水素イオンは触媒と接して分

よれば次のような効果を浸するものである。

- ① 転炉ガスの格製設機からのオフガスより電力, 蒸気,燃料ガス等を一挙に取得でき、転炉ガスの 高付加価値化が可能である。
- ② 電力、蒸気、燃料等の多面的需要に対処できるので、製鉄所周辺の設備合理化が計られる。

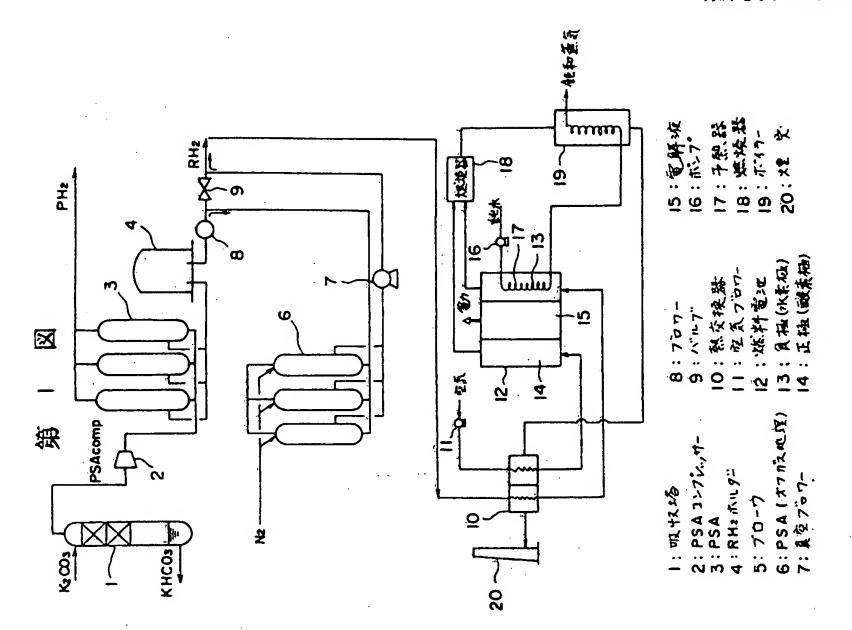
#### 4. 图面の西埠及設明

第 1 図は本発明方法の工程説明図、第 2 図は従来法における工程説明図である。

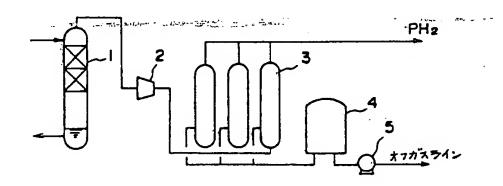
図において 1: 吸収塔、 2: PSAョンブレッサー、 3: PSA、 4: RH。ホルダー、 6: PSA(オフガス処理)、 7: 真空ブローワー、 8: ブローワー、 9: バルブ、 1 0: 熱交換器、 1 1: 空気ブローワー、 1 2: 燃料電池、 1 3: 負極(水素板)、 1 4: 正極(酸染極)、 1 5: 電解液、 1 7: 予熱器、 1 8: 燃烧器、 1 9: ボィラーである。

なお、各図中同一符号は同一または相当部分を 示す。

代理人 弁理士 佐 藤 正 年



### 第 2 図



- 19. Japan Patent Office (JP)
- 12. Laid-open Patent Application Gazette (A)
  - 11. Laid-open Patent Application No. Sho 62-278770
  - Disclosure Date: December 3, 1987
  - 51. Int. Cl.<sup>4</sup> ID Code JPO Control No.

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Examination: Not Requested Yet

Total 4 pages

54. Title of Invention: Method of Recovering CO2 and H2 from

Converter Gas

- 21. Application No. Sho 61-120218
- 22. Application Date: May 27, 1986
- 72. Inventor: Shirou Fujii, Nippon Kokan Co., Ltd., 1-1-2

Marunouchi, Chiyoda-ku, Tokyo

71. Applicant: Nippon Kokan Co., Ltd., 1-1-2 Marunouchi,

Chiyoda-ku, Tokyo

74. Agent: Masatoshi Sato, Patent Agent, and two others

#### SPECIFICATION

- 1. TITLE OF INVENTION
  - Method of Recovering  ${\rm CO_2}$  and  ${\rm H_2}$  from Converter Gas
- 2. WHAT IS CLAIMED IS:

A method of recovering CO<sub>2</sub> and H<sub>2</sub> from converter gas in which converter gas is modified, and then the CO<sub>2</sub> in the modified gas is absorbed in an absorption tower and CO<sub>2</sub> is obtained from the absorption solution, and then the absorption tower offgas is pressurized and the pressurized gas is treated by a PSA (pressure swing adsorber), wherein PSA offgas is additionally treated by a PSA installed in series and then supplied to a fuel cell, with electricity obtained and the fuel cell completely combusting the offgas, and the combustion heat is converted to steam and utilized.

#### 3. DETAILED DESCRIPTION OF THE INVENTION

[Industrial Field Of Application]

The present invention pertains to a method of recovering  ${\rm CO}_2$  and  ${\rm H}_2$  from converter gas generated from an LD converter, etc. when making steel and obtaining heat and electrical energy.

#### [Prior Art]

In recent years, LD converters, etc. used for making steel have sought to actively recover and effectively utilize the converter exhaust gas, which contains CO,  $CO_2$ , and  $H_2$  generated during operations.

These converter gas components, on a volume standard (hereinafter the same) are  $CO_2$  10~15%,  $H_2$  1%, CO 65~75%,  $N_2$  10%, and  $O_2$  1%.

This converter gas itself can be used as fuel gas, but sometimes a recovery method is performed in which this gas is used as a raw-material gas, with steam added to it to cause a shift reaction that makes carbon dioxide and hydrogen; this increases the added value.

FIG. 2 is a process diagram of the conventional method of recovering  ${\rm CO_2}$  and  ${\rm H_2}$  from LD converter gas.

In the drawing, LD converter gas with the aforesaid composition is used as a raw material and passes through a modification step in which this gas is pressurized to  $9~20~{\rm kg/cm^2}$ , steam is added to it, a shift reaction is caused, and the gas components are modified. (More than this is not shown in the drawing.)

Next, the modified gas is introduced to an absorption tower 1, the  $\mathrm{CO}_2$  in the modified gas is absorbed in the potassium carbonate solution ( $\mathrm{K}_2\mathrm{CO}_3$ ) that is the absorption solution and makes potassium bicarbonate (KHCO $_3$ ); this is stripped with steam and the absorption solution becomes  $\mathrm{K}_2\mathrm{CO}_3$  while the stripped  $\mathrm{CO}_2$  gas is made into a product.

The CO<sub>2</sub> is discharged, and the H<sub>2</sub>-rich offgas in the absorption tower 1 has its pressure increased from 9~20 kg/cm<sup>2</sup> to 9~25 kg/cm<sup>2</sup> by a PSA compressor 2, and is treated by a PSA (Pressure Swing Adsorption device) 3. Gases other than hydrogen are adsorbed in the PSA by utilizing the pressure differential, and separated into product high-

purity hydrogen ( $PH_2$ ) and offgas ( $RH_2$ ). The offgas is stored in an  $HR_2$  holder 4, and is exhausted as fuel for another process by a blower 5.

An example of the components of the offgas in this case are  $CO_2$  12%,  $H_2$  58%, CO 3.5%,  $N_2$  26%, and  $O_2$  0.5%. [Problems the Invention Is to Solve]

As noted above, even though offgas in the conventional recovery method for recovering  ${\rm CO_2}$  and  ${\rm H_2}$  from converter gas contains about 60%  ${\rm H_2}$ , it is merely to be used for fuel.

Hospitals, offices, etc. in the vicinity of a steel plant need steam, electricity, hot water, heating and cooling facilities, etc. Currently, as described previously, even though the CO<sub>2</sub>, H<sub>2</sub>, etc. recovered from converter gas are a beneficial energy source, there are many cases in which the demand of the aforesaid sort of public welfare and management facilities is met by purchased electricity, and heavy oil, etc. is used for hot water and heating and cooling.

The object of the present invention is to provide a method of efficiently recovering CO<sub>2</sub> and H<sub>2</sub> from converter gas that is relatively low in calories (500~1700 Kcal/Nm<sup>3</sup>) for manufacturing equipment but high in hydrogen concentration, and of efficiently utilizing the energy contained therein.

[Means for Solving the Problems]

The present inventors focused on PSA offgas, which contains about 60% hydrogen in the conventional method, studied efficient utilization of this offgas, and arrived at the present invention. That is, the present invention is a method of separating and recovering H<sub>2</sub> from converter gas in which converter gas is modified, and then the CO<sub>2</sub> component in the modified gas is absorbed in K<sub>2</sub>CO<sub>3</sub> solution in an absorption tower and product CO<sub>2</sub> is obtained from the absorption solution, and then the absorption waste gas is pressurized and the pressurized gas is treated by a PSA; it is characterized in that PSA offgas is additionally treated by a PSA installed in series and then the gas (a gas with little CO component and a high-concentration H<sub>2</sub> component)

Japanese → English Laid-open Patent Application No. Sho 62-278770

Klarquist, Sparkman, LLP Ref. No. 6454-58227

is supplied to a fuel cell, electricity is obtained, the fuel cell completely combusts the offgas, and the combustion heat is converted to steam and utilized.

[Operation]

In the inventive method of recovering  $CO_2$  and  $H_2$  from converter gas the PSA offgas, which contains about 60% hydrogen, is additionally treated by a PSA installed in series, and it is possible to efficiently recover high-concentration  $H_2$  with a low CO component, so it is possible to supply it as fuel gas for a fuel cell.

Furthermore, the fuel cell completely combusts the offgas and its exhaust heat can be recovered as steam, so it is possible to advantageously recover in a cost-effective manner various types of energy-electricity, steam, fuel gas, etc.—all at once.

Next, embodiments shall be described.
[Embodiments]

FIG. 1 is a process explanatory diagram for inventive method of recovering  $CO_2$  and  $H_2$  from converter gas.

In FIG. 1, 1~4 are the same as in the aforesaid FIG. 2, so explanation thereof is omitted.

Item 6 is a PSA installed in series, 7 is a vacuum blower, 8 is a blower, 9 is a valve, 10 is a heat exchanger, 11 is an air blower for a combustion, 12 is a fuel cell, 13 is a hydrogen electrode chamber (negative electrode), 14 is an oxygen electrode chamber (positive electrode), 15 is electrolyte, 16 is a pump, 17 is a preheater, 18 is a combustor, 19 is a boiler, and 20 is a chimney.

The inventive method shall be described based on FIG.

1.

The raw-material gas was LD converter gas ( $H_2$ : 0.7%, CO: 69.8%,  $CO_2$ : 14.7%,  $N_2$ : 14.3%) at 1.298  $Nm^3/H$ . This gas was pressurized to 9~11 kg/cm<sup>2</sup> by a compressor, and then this gas was modified to hydrogen through a shift reaction using steam, and then the  $CO_2$  in the gas was absorbed in  $K_2CO_3$  solution, the absorption solution was stripped with steam, and product  $CO_2$  was obtained. Next, the absorption tower 1's offgas was pressurized to 16 kg/cm<sup>2</sup> by the PSA

compressor 2, and treated by the PSA 3.

In the PSA 3, gases other than  $\mathrm{H}_2$  were adsorbed by adsorbent in the tower, and [the offgas] was separated into product  $\mathrm{H}_2$  and offgas  $\mathrm{RH}_2$  containing gases other than  $\mathrm{H}_2$ .

The treated hydrogen gas from the PSA 6 (553  $Nm^3/H$ ) was treated by the PSA 6, so it contains  $H_2$  89.5%, CO 1.1%, CO<sub>2</sub> 0.4%, and  $N_2$  9%; it is something that can be supplied to a fuel cell.

Next, an embodiment for recovering electricity and steam from the fuel cell shall be described.

The treated hydrogen gas from the aforesaid PSA 6 was sucked in by the vacuum blower 7, and at the heat exchanger 10, the offgas temperature was heat-exchanged with waste gas after recovering combustion exhaust heat from the fuel cell 12's offgas and raised to 90°C, and introduced to the hydrogen electrode (negative electrode) of the fuel cell 12.

Japanese  $\rightarrow$  English Laid-open Patent Application No. Sho 62-278770 Klarquist, Sparkman, LLP Ref. No. 6454-58227

Meanwhile, air 2000 nm<sup>3</sup>/H was supplied to the heat exchanger 10 by the blower 11 and heated to 120°C; this air was introduced to the fuel cell 12's oxygen electrode 14.

The fuel cell 12 uses an aqueous phosphoric acid solution 15 as its electrolyte, and has two electrodes made of platinum and ruthenium surrounding this.

At the negative electrode (hydrogen electrode 13), the introduced oxygen gas passes through tiny holes in the electrode, comes close to the reaction zone, is adsorbed by catalyst added to the electrode, and becomes active atomic hydrogen. This atomic hydrogen reacts with hydroxide ions in the electrolyte as in the following equation and becomes water, and when doing so transports two electrons to the electrode.

 $H_2 + 2OH^- -> 2H_2O + 2e^-$ 

At the positive electrode (oxygen electrode 14), the two electrons from the electrode are received, based on the presence of the catalyst, and oxygen molecules in the air supplied from outside react with water from the electrolyte and generate a hydrogen peroxide ion and a hydroxide ion as

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in the following equation. The hydrogen peroxide ion comes into contact with the catalyst and breaks down and forms a hydroxide ion and oxygen. This oxygen is utilized in the electrode reaction once again. Therefore, the positive electrode reaction is:

 $1/2 O_2 + H_2O + 2e^- -> 2OH^-$ 

The hydroxide ions generated by this positive electrode 14 are transported in the electrolyte 15 and reach the negative electrode 13. This formed an entire circuit and produced 740 KW of electricity.

The 190°C offgasses from both the negative electrode 13 and positive electrode 14 of the fuel cell were introduced to the combustor 18 and completely combusted. The 430°C combustion exhaust gas [was supplied to] the boiler 19, where its waste heat was recovered and 6 kg/cm<sup>2</sup> of saturated steam 0.7T/H was recovered.

Furthermore, a preheater 17 for pure water was provided inside the negative electrode 13's chamber, and after preheating [this water] was supplied to the boiler 19.

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The boiler 19's offgas [was supplied to] the heat exchanger 10 and [used to] preheat the gas supplied to the fuel cell and air as described earlier and then vented from a chimney 20.

As a result, the value of converter gas was increased 1.86 times compared to the conventional method.

[Effect of the Invention]

The following sorts of effects are achieved by the inventive method of recovering  ${\rm CO}_2$  and  ${\rm H}_2$  from converter gas.

- (1) Electricity, steam, fuel gas, etc. can be obtained all at once from offgas from converter gas purification equipment, and it is possible to give converter gas higher added value.
- (2) It is possible to respond to a varied demand for electricity, steam, fuel gas, etc., so one can rationalize the facilities in the vicinity of a steel mill.

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#### BRIEF DESCRIPTION OF THE DRAWINGS

4.

FIG. 1 is a process explanatory drawing for the inventive method. FIG. 2 is a process explanatory drawing for the conventional method.

In the drawings, 1 is an absorption tower, 2 is a PSA compressor, 3 is a PSA, 4 is a RH<sub>2</sub> holder, 6 is a PSA (offgas treatment), 7 is a vacuum blower, 8 is a blower, 9 is a valve, 10 is a heat exchanger, 11 is an air blower, 12 is a fuel cell, 13 is a negative electrode (hydrogen electrode), 14 is a positive electrode (oxygen electrode), 15 is electrolyte, 17 is a preheater, 18 is a combustor, and 19 is a boiler.

Furthermore, codes that are the same in both drawings indicate equivalent parts.

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FIG. 1

1: Absorption tower

2: PSA compressor

3: PSA

4: RH<sub>2</sub> holder

5: Blower

6: PSA (offgas treatment)

7: Vacuum blower

8: Blower

9: Valve

10: Heat exchanger

11: Air blower

12: Fuel cell

13: Negative electrode (hydrogen electrode)

14: Positive electrode (oxygen electrode)

15: Electrolyte

16: Pump

17: Preheater

18: Combustor

19: Boiler

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20: Chimney

FIG. 2

\* \* \*

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